LOW TEMPERATURE VISCOSITY IN ELONGATED FERROFLUIDS

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We have studied the relaxation and transport properties of a ferrofluid in an elongational flow. These properties are influenced by the bistable nature of the potential energy. Bistability comes from the irrotational character of the flow together with the symmetry of the dipoles. Additionally, the presence of a constant magnetic field destroys the symmetry of the potential energy magnetizing the system. We have shown that at a moderate temperature, compared to the height of the energy barrier, the viscosity decreases with respect to the value it would have if the potential were stable. This phenomenon is known as the 'negative viscosity' effect. Thermal motion induces jumps of the magnetic moment between the two stable states of the system leading to the aforementioned lowered dissipation effect.

I. INTRODUCTION

The existence of a 'negative' viscosity in dispersions of ferromagnetic particles in a nonpolar solvent is a curious phenomenon which has recently been discovered. It was predicted by Shliomis and Morozov[1] and corroborated experimentally by Bacri *et al.*[2]. Its interest is based upon the fact that contrary to what one would expect, the viscosity of the dispersion as a whole diminishes when an applied magnetic field oscillates at an optimum frequency. As has been known for a long time, the presence of a constant magnetic field prevents free rotation of the dipoles which originates a resistance on the flow of the fluid which increases dissipation. On the other hand, Bacri *et al.*[2] have shown that the increment in the viscosity due to the rotational degrees of freedom is proportional to the difference between the vorticity and the angular velocity of the particles in suspension. Thus, a magnetic field oscillating at a high enough frequency can impart an angular velocity to the particles greater than the vorticity, leading to a 'negative' viscosity. One see that the particles gain kinetic energy at the expense of the oscillating field.

This striking phenomena is the motivation for undertaking this investigation. We have found that the effect described is more general than was thought, considering that it can be applied to any ferrofluid that has a frequency different from the inverse of the Brownian relaxation time.

The key point in our results comes from the fact that we work with axisymmetric particles immersed in a suctioning current. Thus, at a low enough temperature, due to thermal agitation, the particles jump back and forth along the axis of the flow, which thereby introduces an internal frequency in the system. This internal frequency plays exactly the same role as the frequency of the alternating magnetic field in the systems studied by Shliomis and Morozov[1] and Bacri *et al.*[2].

There are examples of systems other than ferrofluids that potentially can show this phenomenology, these are suspensions of gravity dipoles[3,4] or suspensions of magnetotactic bacteria[5]. These bacteria, some of them rod shaped, undergo the phenomenon of magnetotaxis. The bacteria contain iron particles which impart a magnetic moment to themselves.

The paper is organized as follows; in section II we describe the system and perform an analysis of the stability of the equilibrium orientations of the dipoles. Section III is devoted to the derivation of the relaxation equation for the magnetization. In section IV we introduce fluctuations in our analysis and, by applying fluctuating hydrodynamics in the space of orientations, we derive the Langevin equation for the magnetic moment. In section V we compute the stress tensor and the viscosity tensor. Finally, in section VI, we discuss our main results.

II. THE MAGNETIC ROTOR

The system we want to study consists of a dilute colloidal suspension of ferromagnetic rodlike particles immersed in a nonpolar fluid phase. This suspension flows in an elongational flow and under the influence of a constant magnetic

field $\vec{H}$ oriented in the direction of the symmetry axis of the flow, which will be taken parallel to the z-axis. All of the particles are supposed to have the same magnetic moment

$$\vec{m} = m_s \hat{R}, \tag{1}$$

where $\hat{R}$ is the unit vector along the direction of the axis of the particle and m_s is the magnetic moment strength. The velocity field of the flow is given by

$$\vec{v} = \vec{\vec{\beta}} \cdot \vec{r}, \tag{2}$$

where $\vec{\vec{\beta}}$ is the velocity gradient of the flow

$$\vec{\vec{\beta}} = \beta \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 2 \end{pmatrix} \tag{3}$$

with β being the rate of elongation.

A suspended particle in the carrier fluid experiences the hydrodynamic torque given by

$$\vec{T}^{th} = -\vec{\vec{\xi}} \cdot (\vec{\omega} - \vec{\Omega}), \tag{4}$$

where the friction tensor $\vec{\vec{\xi}}$ is given in terms of its components ξ_0 and ξ_1

$$\vec{\vec{\xi}} = \xi_1 \hat{R}\hat{R} + \xi_0 (\vec{\vec{1}} - \hat{R}\hat{R}). \tag{5}$$

Here $\vec{\omega}$, $\vec{\Omega}$ and $\vec{\vec{1}}$ are the angular velocity of the particle, the drag angular velocity due to the motion of the fluid and the unit tensor, respectively. Due to the fact that the only possible relative motion between the ends of the rod is a rigid rotation one has

$$\vec{v}_+ - \vec{v}_- = \vec{\Omega} \times \hat{R}L, \tag{6}$$

where $\vec{v}_\pm$ are the velocities of the end points of the rod and L is its length. This expression can alternatively be written as

$$\vec{v}_+ - \vec{v}_- = \vec{\vec{\beta}} \cdot \hat{R}L. \tag{7}$$

Thus, through a comparison of eqs. (6) and (7) we achieve

$$\vec{\Omega} = \vec{\mathcal{R}}(1/2\vec{\vec{\beta}} : \hat{R}\hat{R}), \tag{8}$$

with $\vec{\mathcal{R}} \equiv \hat{R} \times \frac{\partial}{\partial \hat{R}}$ being the rotational operator.

In addition to eq. (4) the dipole is acted upon by a magnetic torque

$$\vec{T}^m = \vec{m} \times \vec{H}. \tag{9}$$

Therefore, by taking into account the angular momentum conservation, in view of eqs. (4), (5), (8), and (9) we can write in the high friction limit

$$\vec{\omega} = \frac{1}{\xi_0} (-\vec{\mathcal{R}}U), \tag{10}$$

where the potential U, defined through

$$U = -\vec{m} \cdot \vec{H} - \frac{1}{2}\xi_0 \vec{\vec{\beta}} : \hat{R}\hat{R}; \tag{11}$$

accounts for the different mechanisms for the rotation of the particle: the magnetic field and the external flow. In polar spherical coordinates this energy can be rewritten as

$$U = -m_s H \cos\theta + \frac{\xi_0 \beta}{2}(1 - 3\cos^2\theta), \tag{12}$$

θ being the angle between the axis of the particle and the z-axis.

The equilibrium states of the system can be identified through the condition $\frac{dU}{d\theta} = 0$, i.e.

$$m_s H \sin\theta + 3\xi_0 \beta \sin\theta \cos\theta = 0 \tag{13}$$

whose solutions, θ_-, θ_+ and θ_0, are

$$\theta_- = 0, \tag{14}$$

$$\theta_+ = \pi, \tag{15}$$

$$\theta_0 = \arccos\left(-\frac{m_s H}{3\xi_0 \beta}\right). \tag{16}$$

The stability of these orientations follows from the second derivative of the potential computed in each of the solutions

$$\frac{d^2U}{d\theta^2}\Big|_{\theta_-} = m_s H + 3\xi_0 \beta = m_s H(1 + \frac{H_c}{H}), \tag{17}$$

$$\frac{d^2U}{d\theta^2}\Big|_{\theta_+} = -m_s H + 3\xi_0 \beta = m_s H(\frac{H_c}{H} - 1), \tag{18}$$

$$\frac{d^2U}{d\theta^2}\Big|_{\theta_0} = \frac{(m_s H)^2}{3\xi_0 \beta} - 3\xi_0 \beta. \tag{19}$$

Since m_s, H, ξ_0 and β are always positive quantities, after examining eqs. (17)-(19) we conclude that provided

$$H \leq H_c, \tag{20}$$

$H_c \equiv 3\xi_0 \beta / m_s$ being a critical field, θ_- and θ_+ are both stable, whereas θ_0 is unstable. We then conclude that under these conditions the potential of the magnetic rotor is bistable. For magnetic fields larger than H_c the system becomes stable.

Equivalently, we could have considered the presence of a critical value for the elongational rate, $\beta_c = \frac{H m_s}{3\xi_0}$, such that for a fixed value of the magnetic field the system is bistable whenever the actual value of the elongational rate β overcomes β_c. In the rest of the paper we will assume that $H/H_c < 1$, i. e. we will remain in the bistable region, or in the range of large elongational rate.

To conclude this section we will give some estimates of the critical field corresponding to situations of experimental accesibility. For particles of magnetite having a volume $V_p = 5 \times 10^{-19} cm^3$ and an aspect ratio $\epsilon = 0.1$ one has $m_s = 2.4 \times 10^{-16} G \times cm^3$, and consequently $H_c = 58.3 Oe$. On the other hand, for particles of cobalt with volume $V_p = 2.7 \times 10^{-19} cm^3$ and $\epsilon = 0.1$ we obtain $m_s = 3.8 \times 10^{-16} G \times cm^3$, and $H_c = 17.1 Oe$. In both cases we have assumed $\beta = 10^3 s^{-1}$. At smaller elongational rate, $\beta = 10 s^{-1}$, one has $H_c = 5.8 Oe$ for magnetite and $H_c = 1.7 Oe$ for cobalt.

III. RELAXATION EQUATION

The relaxation of the magnetic rods can be interpreted as a diffusion process through a potential barrier in orientation space. In this scenario we can apply the formalism of non-equilibrium thermodynamics[6]. Let $\rho(\hat{R}, t)$ be the angular distribution function that may be viewed as a density in the space of orientations. Thus, we can define the chemical potential as

$$\mu = K_B T \ln\rho + U. \tag{21}$$

The diffusion equation for ρ is written in the form

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$$\frac{\partial \rho}{\partial t} = \frac{D}{\sin \theta} \frac{\partial}{\partial \theta} \{\sin \theta [\frac{\rho}{K_B T} \frac{\partial U}{\partial \theta} + \frac{\partial \rho}{\partial \theta}]\}, \tag{22}$$

with $D = k_B T / \xi_0$ and where the axial symmetry of the potential (12) has been taken into account. Equation (22) can be rewritten as a continuity equation

$$\frac{\partial \rho}{\partial t} = -\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} (J_\theta \sin \theta), \tag{23}$$

which defines the diffusion current

$$J_\theta = -D e^{(-U/K_B T)} \frac{\partial}{\partial \theta} e^{\mu/K_B T}. \tag{24}$$

If the height of the potential barrier is large enough as compared to thermal energy $K_B T$, we can suppose that equilibrium is reached independently on each side of the barrier. Thus, the chemical potential can be inferred as being

$$\mu(\hat{R}, t) = \mu(\theta_-) \Theta(\theta_0 - \theta) + \mu(\theta_+) \Theta(\theta - \theta_0), \tag{25}$$

where $\Theta(\theta)$ is the unit step function. The system is allowed to obtain the global equilibrium due to the presence of a quasi-stationary current $J(t)$, assumed uniform.

$$J_\theta \sin \theta = J(t) \{\Theta(\theta - \theta_-) + \Theta(\theta - \theta_+)\}. \tag{26}$$

This adiabatic hypothesis is one of the essential points in the present development. By using (21) and (25), one has

$$\rho(\hat{R}, t) = \rho_-(t) e^{[-(U - U_-)/K_B T)]} \Theta(\theta_0 - \theta) + \rho_+(t) e^{[-(U - U_+)/K_B T)]} \Theta(\theta - \theta_0), \tag{27}$$

where $\rho_\pm \equiv \rho(\theta_\pm, t)$, and $U_\pm \equiv U(\theta_\pm)$, are the densities and potential energies in the two stable states. In order to obtain an expression for $J(t)$, we substitute (24) into (26) to arrive at

$$J(t) \frac{e^{U/K_B T}}{\sin \theta} \{\Theta(\theta - \theta_-) + \Theta(\theta - \theta_+)\} = -D \frac{\partial}{\partial \theta} e^{\mu/K_B T}. \tag{28}$$

Integrating now over θ and taking into account that, due to the height of the barrier, the main contribution to these integrals is around the maximum of the potential θ_0, one has the law of mass action[7]

$$J(t) = K_B l (1 - e^{A/K_B T}), \tag{29}$$

which is a nonlinear phenomenological relationship between the quasi-stationary current $J(t)$ and the affinity $A \equiv \mu_+ - \mu_-$, where $\mu_\pm \equiv \mu(\theta_\pm)$. The phenomenological coefficient l is given by

$$l = \frac{D \sin \theta_0}{K_B} \rho_- (\frac{|U_0''|}{2\pi K_B T})^{1/2} e^{(U_0 - U_-)/K_B T}, \tag{30}$$

where $U_0'' \equiv \frac{d^2 U}{d\theta^2}|_{\theta_0}$ and $U_0 \equiv U(\theta_0)$.

We are now prepared to proceed to the deduction of the relaxation equations. To this end, we define the following populations:

$$N_+ \equiv \int_{\phi=0}^{2\pi} \int_{\theta=\theta_0}^{\pi} \rho(\hat{R}, t) d\hat{R} = \int_{\theta_0}^{\pi} 2\pi \sin \theta \rho(\hat{R}, t) d\theta, \tag{31}$$

and

$$N_- \equiv \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\theta_0} \rho(\hat{R}, t) d\hat{R} = \int_0^{\theta_0} 2\pi \sin \theta \rho(\hat{R}, t) d\theta, \tag{32}$$

related by the normalization condition

$$N = N_+ + N_-. \tag{33}$$

The kinetic equations for N_+ and N_- follow after differentiating the eqs. (31) and (32) and employing eq. (23). Thus, we have

$$\frac{dN_+}{dt} = -\frac{dN_-}{dt} = 2\pi J(t), \tag{34}$$

where, consistently with the adiabatic approximation

$$N_- = \frac{2\pi K_B T}{U''_-}\rho_-, \tag{35}$$

$$N_+ = \frac{2\pi K_B T}{U''_+}\rho_+, \tag{36}$$

in which $U''_\pm \equiv \frac{d^2 U}{d\theta^2}|_{\theta_\pm}$. Using equations (29) and (34)-(36) we can derive the rate equations for the two populations

$$\frac{dN_+}{dt} = -\frac{dN_-}{dt} = K_{+-}N_- - K_{-+}N_+, \tag{37}$$

where the rate constants are given by

$$K_{+-} = \frac{D \sin\theta_0}{2\pi K_B T}(\frac{|U''_0|}{2\pi K_B T})^{1/2}U_- e^{-(U_0-U_-)/K_B T}, \tag{38}$$

and

$$K_{-+} = \frac{D \sin\theta_0}{2\pi K_B T}(\frac{|U''_0|}{2\pi K_B T})^{1/2}U''_+ e^{-(U_0-U_+)/K_B T}. \tag{39}$$

We can now define the magnetic moment parallel to the direction of the applied magnetic field as

$$m \equiv m_s \frac{N_+ - N_-}{N}. \tag{40}$$

By differentiating equation (40) and employing equation (37) we obtain the relaxation equation for m

$$\frac{dm}{dt} = -\frac{1}{\tau}m + \frac{1}{\alpha}m_s, \tag{41}$$

where

$$\tau = \frac{1}{K_{+-} + K_{-+}} \tag{42}$$

is the relaxation time, and

$$\alpha = \frac{1}{K_{+-} - K_{-+}} \tag{43}$$

accounts for the asymmetry of the potential. An equation similar to (41) was postulated by Shliomis in the context of ferrohydrodynamics[8]. Our approach to deduce eq. (41) has been based upon mesoscopic arguments and as we will see in the next section it provides the natural way of introducing fluctuations in the scheme.

It is useful for our purposes to introduce the nondimensional variables $x \equiv \frac{H_c}{H}$ and $\mu \equiv \frac{m_s H}{K_B T}$, in terms of which the relaxation time is written as

$$\tau = \frac{(2\pi)^{3/2}}{D}(x\mu)^{(-1/2)}(1 - \frac{1}{x^2})^{-1}\exp\{\frac{\mu}{2}(\frac{1}{x}+$$

$$\frac{x}{3})\}[\mu(x-1)\exp\{\mu(1-\frac{x}{3})\} + \mu(x+1)\exp\{-\mu(1+\frac{x}{3})\}]^{-1}, \tag{44}$$

where x must be greater than the unity. The inverse of τ gives us a characteristic frequency proper to the system, the jump frequency between the two stable states of the potential energy. This frequency is given by

$$\omega = D(\frac{x\mu}{2\pi})^{3/2}(1 - \frac{1}{x^2})e^{-\frac{x\mu}{2}}e^{-\mu/x}[e^\mu + e^{-\mu} + \frac{1}{x}(e^{-\mu} - e^\mu)]. \tag{45}$$

In Figure 1 we can observe the behavior of ω when one varies the elongational rate. The way in which it depends on the elongational rate and the existence of two time scales in our problem, Brownian D^{-1} and τ, are the key points in understanding the dynamical mechanism which leads to the results we will obtain in section 5.

IV. DYNAMIC OF THE FLUCTUATIONS OF THE MAGNETIC MOMENT

Thermal motion of the dipoles inside the carrier fluid produces fluctuations in the population of the minima of the potential. This fact manifests itself on a mesoscopic level through fluctuations of the magnetic moment. These can be taken into account by adding a random current[6] to equation (29),

$$J(t) = K_B l(1 - e^{A/K_B T}) + \frac{\sqrt{\bar{D}}}{m_s}\xi(t), \tag{46}$$

where $\bar{D}$ is given by

$$\bar{D} = 2m_s^2 k_B \bar{l}, \tag{47}$$

$\bar{l}$ is the equilibrium value of the phenomenological coefficient l, which according to (30) should be proportional to ρ_- computed at equilibrium, ρ_-^{eq}. By applying the detailed balance principle to eq. (37) and using the normalization relation eq. (33) one can easily achieve

$$\rho_-^{eq} = \left(\frac{N}{2\pi k_B T}\right)\frac{U_-'' U_+''}{U_+'' + U_-'' \exp\{\frac{U_- - U_+}{k_B T}\}}. \tag{48}$$

Additionally, in eq. (46) $\xi(t)$ is a Gaussian white noise stochastic process of zero mean and correlation function.

$$\langle\xi(t)\xi(t')\rangle = \delta(t - t'). \tag{49}$$

From equations (34), (40) and (46) we obtain the Langevin equation for m

$$\frac{dm}{dt} = -\frac{1}{\tau}m + \frac{1}{\alpha}m_s + \sqrt{\bar{D}}\xi(t). \tag{50}$$

Notice that coefficient $\bar{D}$ is the input noise strength corresponding to the stochastic process m. This coefficient can by explicitly written as

$$\bar{D} = m_s^2 D\sin\theta_0(\frac{|U_0''|}{2\pi K_B T})^{1/2}(1 + \frac{\tau}{\alpha})\frac{U_+''}{2\pi K_B T}exp\{-\frac{(U_0 - U_+)}{K_B T}\} \tag{51}$$

and in terms of the nondimensional parameters x and μ

$$\bar{D} = \frac{m_s^2 D}{(2\pi)^{3/2}}(x\mu)^{1/2}(1 - \frac{1}{x^2})\mu(x-1)\frac{2(1+x)e^{-\mu}}{(1+x)e^{-\mu} + (x-1)e^\mu}\exp\{-\frac{\mu}{2}(\frac{1}{x} + \frac{x}{3})\}exp\{\mu(1 - \frac{x}{3})\}. \tag{52}$$

Finally, in reference to eq. (50), by performing the change

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$$\tilde{m} = m - \frac{\tau}{\alpha} m_s, \tag{53}$$

this equation becomes

$$\frac{d\tilde{m}}{dt} = -\frac{1}{\tau}\tilde{m} + \sqrt{\bar{D}}\xi(t), \tag{54}$$

i.e., $\tilde{m}$ is an Ornstein-Uhlenbeck process[9]. As is well known, the stationary distribution for such a process is

$$p(\tilde{m}) = \frac{1}{\sqrt{\pi\tau\bar{D}}} \exp\{-\frac{\tilde{m}^2}{\tau\bar{D}}\}, \tag{55}$$

which will be used in the next section to compute the viscosity.

V. THE VISCOSITY TENSOR

In order to calculate the viscosity tensor, we first have to compute the stress tensor. This quantity has two contributions, one that comes from the solvent, and the other due to the presence of the particles. The latter is written[10] as

$$\vec{\vec{\sigma}} = nK_BT(3\langle\hat{R}\hat{R}\rangle - \vec{\vec{1}}) + n\xi_1\vec{\vec{\beta}} : \langle\hat{R}\hat{R}\hat{R}\hat{R}\rangle - nK_BT\mu\langle\hat{R}\hat{H}\cdot(\vec{\vec{1}} - \hat{R}\hat{R})\rangle, \tag{56}$$

where n is the concentration of suspended particles and L is their longitude. Thus, eq. (56) give us first order contributions to the viscosity tensor. The moments in eq. (56) will be calculated by using the stationary distribution (55). It is important to keep in mind that although we are using a stationary distribution, the dynamical effects are taken into account through their dependence on the relaxation time τ.

We will illustrate the behavior of the viscosity by explicitly computing the parallel viscosity defined through

$$\eta_{\|\|\|} = \frac{\sigma_{\|\|\|}}{\beta} \tag{57}$$

with $\sigma_{\|\|\|} = \hat{H}\cdot\vec{\vec{\sigma}}\cdot\hat{H}$, that from eq. (56) is written as

$$\sigma_{\|\|\|} = nK_BT(3\langle\hat{R}_\|\hat{R}_\|\rangle - 1) + n\xi_1\beta(\langle\hat{R}_\|\hat{R}_\|\rangle - \langle\hat{R}_\|\hat{R}_\|\hat{R}_\|\hat{R}_\|\rangle) - nK_BT\mu(\langle\hat{R}_\|\rangle - \langle\hat{R}_\|\hat{R}_\|\hat{R}_\|\rangle). \tag{58}$$

Here we have taken into account that $\vec{\vec{\beta}} : \hat{R}\hat{R} = \beta(1 - 3\hat{R}_\|^2)$, and $\hat{R}_{\|\|\|} = \hat{R}\cdot\hat{H}\hat{H}$. In the appendix we summarize the result of the computation of all the moments that appear in the expression for $\sigma_{\|\|\|}$. Making use of these results, we find that

$$\sigma_{\|\|\|} = nK_BT[\frac{\tau\bar{D}}{2m_s^2} + (\frac{\tau}{\alpha})^2 - 1] + n\xi_1\beta[(\frac{\tau}{\alpha})^2 + \frac{\tau\bar{D}}{2m_s^2} - 9(\frac{\tau\bar{D}}{2m_s^2})^2 -$$

$$18\frac{\tau}{\alpha}\frac{\tau\bar{D}}{2m_s^2} - 3(\frac{\tau}{\alpha})^4] - nK_BT\mu[\frac{\tau}{\alpha} - (\frac{\tau}{\alpha})^3 - 3\frac{\tau}{\alpha}\frac{\tau\bar{D}}{2m_s^2}]. \tag{59}$$

Thus, we finally obtain

$$\frac{\eta_{\parallel\parallel}}{3n\xi_0} = \frac{1}{x\mu}[(1+\mu)\frac{\tau}{\alpha} + \frac{\tau\bar{D}}{2m_s^2} + 3\mu\frac{\tau}{\alpha}\frac{\tau\bar{D}}{2m_s^2} + \mu(\frac{\tau}{\alpha})^3 - 1] +$$

$$\frac{1}{3}\frac{\xi_1}{\xi_0}[\frac{\tau\bar{D}}{2m_s^2} - 18\frac{\tau}{\alpha}\frac{\tau\bar{D}}{2m_s^2} - 9(\frac{\tau\bar{D}}{2m_s^2})^2 + (\frac{\tau}{\alpha})^2 - 3(\frac{\tau}{\alpha})^4]. \tag{60}$$

In figures 2 and 3 we have plotted the nondimensional quantity $\eta_{par} \equiv \frac{\eta_{\parallel\parallel}}{3n\xi_0}$ in terms of x and ω, respectively, for particles with an aspect ratio $\epsilon = 0.1$, for which one has $\frac{1}{3}\frac{\xi_1}{\xi_0} = 0.1805$[11].

One can observe that this viscosity becomes negative for large shear rates and saturates at a positive value. Similar results have been obtained by Shliomis and Morozov[1] and Bacri *et al.*[2] with ferrofluids under an oscillating magnetic field. Negative viscosities in these cases were due to the existence of two characteristic time scales in the system that enter in competition. One of them is related to the frequency of the alternating magnetic field and the other is related to the vorticity of the fluid. In our case, the dipoles relax to the equilibrium orientations in a time scale D^{-1}, which is shorter than the relaxation time τ associated with the diffusion through the potential barrier. The rods are then constantly jumping with a frequency $\omega = \tau^{-1}$ acquiring a net angular velocity different from zero, whose expression is given in the appendix. The existence of this angular velocity then shows the conversion of thermal energy into kinetic energy for rotation. This explains why, in our case, the viscosity diminishes.

In looking at fig. 3 one first observes a histeresys cycle described by η_{par} when ω varies. During the first stage, when the actual value of ω is between zero and its maximum value, η_{par}, decreases from a positive value to a minimum negative value, as occurs in the case analyzed by Shliomis and Morozov[1]. These results are in good agreement with those found by them. The difference between our approach and the one by Shliomis and Morozov[1] is that the externally imposed frequency can be as large as you want. In our case the frequency of relaxation, the result of an internal dynamical mechanism, does not grow freely. In the second stage; ω is a decreasing function of the shear rate. This fact is responsible for the particular behavior of η_{par} observed. Note that, as can be seen in Figure 2, η_{par} does not saturate to a negative value when $\beta \to \infty$. It tends very slowly to a positive value, as slowly as ω tends to zero. Our results also agree with the analysis of the phenomenon carried out by Rosensweig[12] as well.

VI. CONCLUSIONS

In this paper we have shown that in a ferrofluid made from rodlike particles under a constant magnetic field and in a elongational flow, rotations of the particles lead to non-monotonous behavior of the viscosity. This fact allows us to generalize the phenomenon discovered by Bacri *etal.*[2].

As a consequence of the orientating effect of the flow, the particles rotate with a drag angular velocity $\vec{\Omega}$ that eliminates the degeneracy of the direction of their actual angular velocity in a volume element of the ferrofluid (see eq. (10)). Thus, all the contributions to dissipation in a volume element add up constructively.

Moreover, because the flow is elongational, we can write a potential energy of orientation related to it. This potential is bistable and the addition of a magnetic field breaks up its symmetry, thus magnetizing the system. On the other hand, thermal motion causes jumps between the two stable states of the potential with a certain frequency. Consequently, the particles acquire a kinetic energy for rotation at the expense of thermal energy. This fact eliminates the rigidity that the system would have if no motion were present, i.e. if the potential time independent were stable. In fact, this is the way that we interpret the 'negative viscosity' effect predicted for our system.

We have derived the kinetic equations for the population of the minima of the potential and from it the relaxation equation for the magnetic moment. Likewise, by assuming fluctuations of the density of states in the orientation space, we have formulated the Langevin equation for the magnetic moment. This equation describes a Ornstein-Uhlembeck process whose moments are well known. The computation of the first four moments allows us to derive the viscosity.

It should be emphasized that the viscosity is computed in the limit of high energy barrier, i.e. for strong rate of elongation. This is the opposite situation to the case studied in the two previous papers[11,13] where we covered the weak flow regime, showing an increase of the viscosity. Analogously, modifying the magnetic field without altering the rate of elongation we achieve the same effect, that is, to vary the viscosity. Thus, a possible application of our results is in adaptive dampers.

Through this behavior of the viscosity, emerge the macroscopic consecuences of the dynamical bifurcation with exchange of stability that our system experiences.

Likewise, the effect that we are studying is only possible in dilute solutions, where each rod can rotate freely without interference by others. Of course in neglecting hydrodynamic interactions among the particles we assume a extremely dilute ferrofluid. Anyway, this is the first step in our study of nonlinear or hysteresis effects in the rheology of ferrofluids suspensions.

The following step will be to assume higher concentrations. Nonetheless, to include hydrodynamic and exclude volume interactions in the dynamics of an assembly of rod particles is difficult. So that, we will model the elongated dipoles by means of rigid dumbbells, such models are well studied in the field of polymeric liquids.

In order to increase the modulation effect of the magnetic field or of the elongation rate on the viscosity, another possibility we are thinking of consists in adding nonmagnetic spheres at high concentration to the ferrofluid.

APPENDIX A:

To begin with, in this appendix we give the moments which must be calculated in order to compute the stress tensor.

$$\langle \hat{R}_\parallel \rangle = \frac{\tau}{\alpha} \tag{A1}$$

$$\langle \hat{R}_\parallel \hat{R}_\parallel \rangle = \frac{\langle \tilde{m}^2 \rangle}{m_s^2} + (\frac{\tau}{\alpha})^2 = \frac{\tau \bar{D}}{2m_s^2} + (\frac{\tau}{\alpha})^2 \tag{A2}$$

$$\langle \hat{R}_\parallel \hat{R}_\parallel \hat{R}_\parallel \rangle = 3\frac{\tau \bar{D}}{2m_s^2}\frac{\tau}{\alpha} + (\frac{\tau}{\alpha})^3 \tag{A3}$$

$$\langle \hat{R}_\parallel \hat{R}_\parallel \hat{R}_\parallel \hat{R}_\parallel \rangle = 3(\frac{\tau \bar{D}}{2m_s^2})^2 + 6(\frac{\tau}{\alpha})^2 \frac{\tau \bar{D}}{2m_s^2} + (\frac{\tau}{\alpha})^4 \ . \tag{A4}$$

Finally, according to eqs. (8), (10) we can write

$$\vec{\omega} - \vec{\Omega} = \frac{1}{\xi_0}\vec{m} \times \vec{H}. \tag{A5}$$

Now we define the root mean square angular velocity $\omega_{rm} \equiv \sqrt{\langle (\vec{\omega} - \vec{\Omega})^2 \rangle}$, that in view of eq. (A5) is given by

$$\omega_{rm} = \mu D \left\{ 1 - \frac{\tau \overline{D}}{2m_s^2} - \left(\frac{\tau}{\alpha}\right)^2 \right\}^{1/2} . \tag{A6}$$

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FIGURE CAPTIONS

- Figure 1.- Nondimensional frequency versus the scaled elongational rate x, for $\mu = 1$.

- Figure 2.- Normalized viscosity versus the scaled elongational rate x, for $\mu = 1$.

- Figure 3.- Normalized viscosity versus the nondimensional jump frequency for $\mu = 1$.